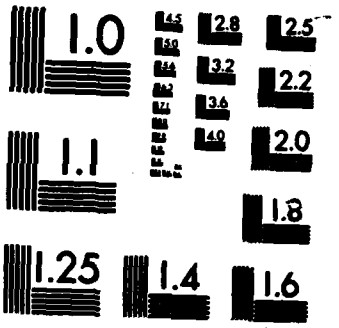
AD-A150 953 SPATIAL RESOLUTION OF SMALL PARTICLES IN SILANE DISCHARGE(U) NORTHWESTERN UNIV EVANSTON IL DEPT OF CHEMISTRY K G SPEARS ET AL. 18 FEB 85 TR-8 1/1

UNCLASSIFIED N00014-79-C-0622 F/G 7/4 NL



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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER: Technical Report No. 8	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle): Spatial Resolution of Small Particles in Silane Discharge		5. TYPE OF REPORT & PERIOD COVERED: Interim
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s): K. G. Spears and R. M. Roth		8. CONTRACT OR GRANT NUMBER(s): N00014-79-0622
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Northwestern University, Department of Chemistry		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: NR 056-729
11. CONTROLLING OFFICE NAME AND ADDRESS: Office of Naval Resarch, Chemistry Program, Code 472, Arlington, VA 22217		12. REPORT DATE: 2/18/85 13. NUMBER OF PAGES: 6
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office): ONR, Chicago, 536 S. Clark, Chicago, IL 60605		15. SECURITY CLASS. (of this report): Unclassified 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

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17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

To be published in a Proceedings of a Symposium on Plasma Synthesis and Etching of Electronic Materials, Materials Research Society, Boston Nov. 1984

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

rf discharge, silane, plasma chemistry, particles, clusters, laser light scattering.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

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DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE S/N 0102-014-6601

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

To be published in *Proceedings of a Symposium on Plasma Synthesis and Etching of Electronic Materials*, Materials Research Society, Boston, Nov. 1984

SPATIAL RESOLUTION OF SMALL PARTICLES IN SILANE DISCHARGE

K. G. SPEARS AND R. M. ROTH*

Northwestern University, Department of Chemistry, Evanston, IL 60201
*Currently at Standard Oil Company (Indiana), Amoco Research Center, P. O. Box 400 Naperville, IL 60566

ABSTRACT

A capacitively coupled, rf glow discharge of silane in argon was studied with laser light scattering to determine the spatial concentration of small particles. Multi-wavelength scattering profiles have been obtained and are being analyzed to obtain size distributions as a function of spatial location. Very sharply defined particle zones can be found under some plasma conditions that are spatially related to the silicon atom profiles. We will report our results and attempt to qualitatively describe how these zones relate to plasma chemistry and film deposition processes.

INTRODUCTION

The existence of particle light scattering in silane plasmas is a recognized phenomenon [1,2,3] that has yet to be understood and correlated with film formation properties. In this paper, we present results from spatially resolved studies of particle light scattering in a capacitively coupled rf glow discharge of silane. We have made an earlier report [4] on our light scattering results and in this paper we will briefly summarize those results and provide new data on particle light scattering and its relation to silicon atom fluorescence signals.

The apparatus is identical to the system described in our accompanying paper [5]. We have used both 500.0 and 251.4 nm radiation in various independent light scattering experiments, but without a direct comparison in the same experiment. The scattered light was detected at 90° through a monochromator and the input polarization optimized the scattered light intensity when it was perpendicular to the plane containing the input and scattered rays. The scattered intensity was a great deal larger than atomic emission from silicon atoms which was also excited simultaneously in some experiments at 251.4 nm. It was possible to use the resolved fluorescence emission line at 252.85 nm to study fluorescence without interference from large light scattering signals at the 251.43 nm excitation. Polarized light scattering could also be rejected by proper polarization. The size of the Brewster angle windows prevented a complete spatial scan between the electrodes when we optimized the laser polarization to maximize the light scatter detection so that we only show scattering for regions near the two electrodes. The ground electrode is labeled at position zero and the rf electrode is labeled at position 22 mm in the following figures.

RESULTS

A typical light scattering result is shown in figure 1. This figure shows light scattering with 500.0 nm radiation of 10 ns duration and a maximum energy of 0.1-0.2 mJ. The focal spot size was ~ 0.1 mm diameter and scattering from the electrodes only occurs in the closest 0.25 mm zone, which is excluded from the following figures. The discharge conditions of figure 1 were 5% SiH_4 in argon, 3 W in power, and a pressure of 0.50 torr. The electrodes have unequal areas because of a ground screen confining the plasma and a bias of -40 V builds up on the cathode. These discharge conditions yield a-Si:H film having typical IR absorbance features.

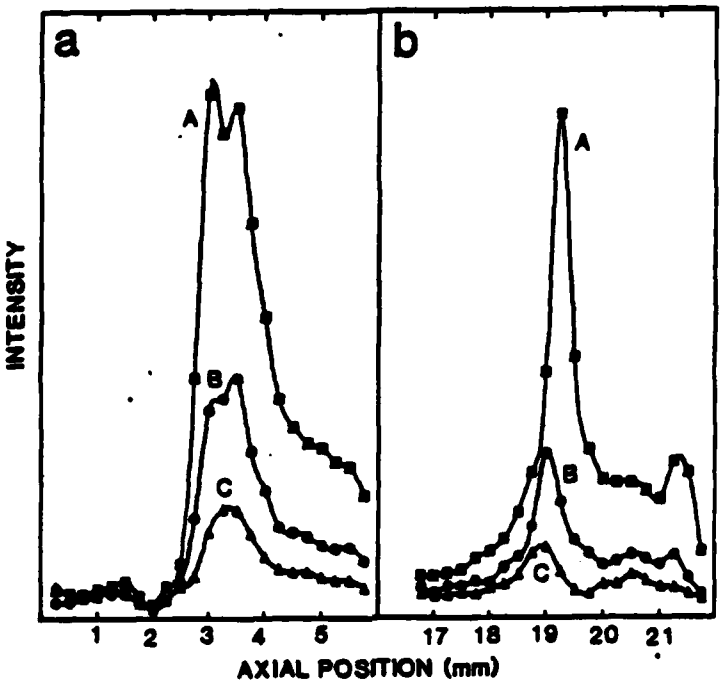

Figure 1: Light scattering signal for 500 nm excitation. Flow rates of silane were: A - 35 sccm, B - 40 sccm, C -45 sccm.

The scattering signals in figure 1 show qualitative differences between the anode (electrical ground) and cathode (rf electrode) for any given flow rate. The intensity of scattering is normalized in figure 1, but the intensity is greater at the cathode. The spatial extent of scattering is much less at the cathode than the anode, and both electrodes show light scattering at close distances (~ 1 mm) from the electrode surfaces. In addition, substructure can be seen in the large peak near the anode. As demonstrated in earlier work [4], the location of the main scattering peaks is quite dependent on the pressure for a given flow rate and mole fraction of SiH_4. This effect shows a correspondence with the expected behavior of plasma sheaths [4,6]. The large light scattering peak at the cathode shows a constant product of electrode displacement distance, d, and the pressure, p, of 1.00 ± .04 mm torr over the range 0.6 to 0.3 torr [4]. We have interpreted this behavior as demonstrating the importance of the ion sheath boundary in defining a zone for particle growth. In this paper we will present more data to further understand the existence of the sharp spatial profiles.

The data in figure 1 show particle scattering for several flow rates at constant pressure and mole fraction. Additional data with flows as low as 25 sccm confirm a dramatic dependence of particle light scattering signal on

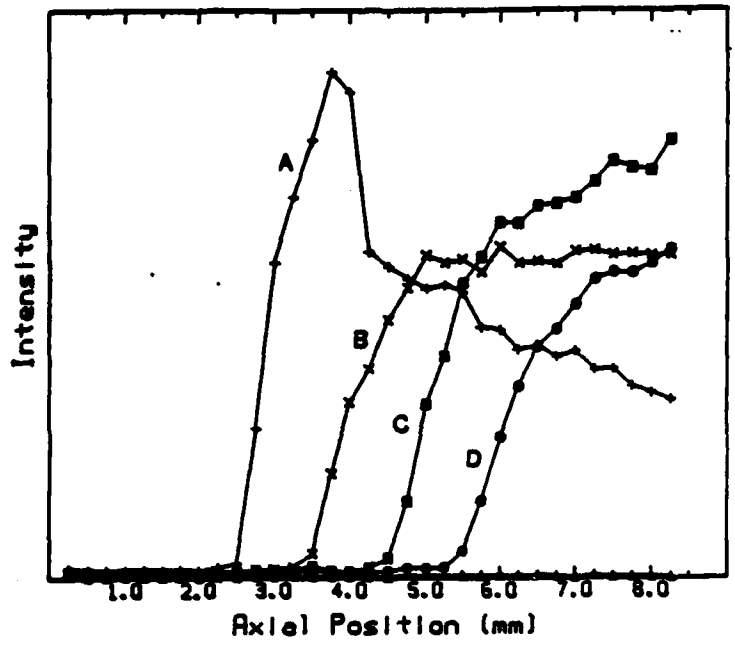

Figure 2: Light scattering for 251.4 excitation. Mole fractions of silane in argon were: A - 0.45%, B - 0.3%, C - 0.2%, D - 0.1%, E - 0.05%. The pressure was 0.50 torr at 50 sccm flow and a power of 3W. The curve E shows no signal above the noise.

flow rate. The light scattering signal increases nearly logarithmically with decreasing flow rate. Flow rate changes also affect the position and intensity of light scattering signals from the smaller peaks closer to the electrode more than the larger peaks.

The origin of sharp, scattering peaks can be further understood by progressively reducing the mole fraction of silane to a limit where no scattering can be observed. One goal of these studies was to identify a perturbation limit where the discharge parameters, such as the electron distribution, are dominated by argon. When silane mole fractions are decreasing from 8 to 0.5%, there is an increase of particle scattering signal with little change in the qualitative feature of peaked light scattering, except that below 2%, the sharpness of the peak is reduced on the side toward the middle of the discharge. The progressive changes at mole fractions below 0.5% are shown in figure 2 for the anode side of the discharge. This data was obtained with 251.4 nm light scattering in order to track Si atoms and scattering in successive scans by merely changing the detection wavelength. The photomultiplier gain for the atom fluorescence signal was 12 times larger than the light scattering signal. The change

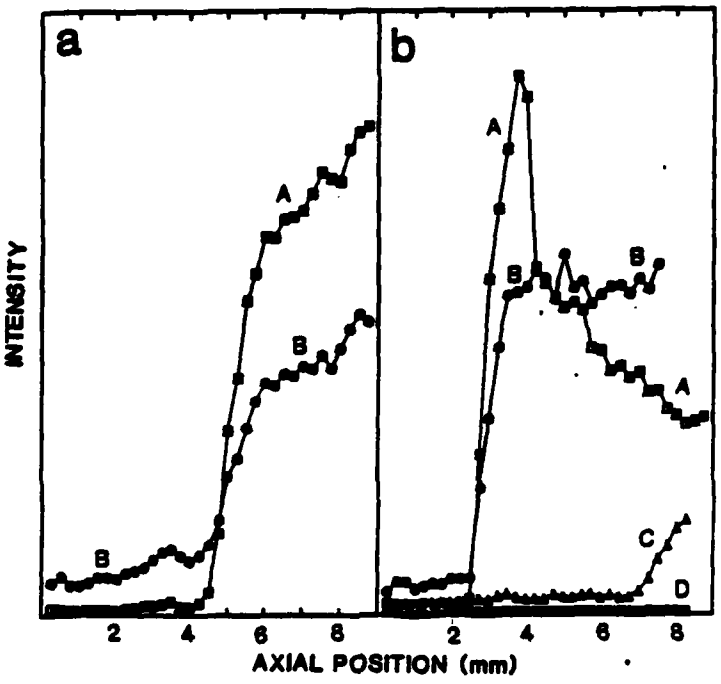

Figure 3: Light scattering and silicon atom fluorescence for 251.4 nm excitation. Conditions as in figure 2. 3a. The light scattering is curve A and atom fluorescence is curve B. Silane mole fraction of 0.2%. 3b. The silane mole fraction of 0.45% has curve A for scattering and B for atom fluorescence. The silane mole fraction of 0.05% has curve D for scattering (none observable) and C for atom fluorescence.

from 0.45% to 0.3% silane shown in figure 2 shows a progressive diminution of the peak (data at 0.4% and 0.35% are not shown). The data for 0.2% and 0.1% silane both show a well defined peak at 3.5 mm that on an expanded scale is ~ 47 times smaller than the peak for 0.45%. The signal at 0.05% silane shows no scattering features, which means that any scattering signal is less than ~ 700 times the large peak for 0.45% (our current noise limit).

The preceding figures reinforce the importance of particles in rf plasmas of silane. The direct relationship of these particles to the creation of silicon atoms was discussed in our accompanying paper [5]. The comparison of spatial profiles of silicon atom concentration and particle light scattering, yields some new insight into these signals. Figure 3a shows light scattering and silicon atom concentration for 0.2% silane. Both signals show a small peak at the 3.5 mm point that is tentatively postulated

as the ion sheath boundary. The 0.3%, 0.2%, and 0.1% curves show very similar behavior for particle scattering and atomic signals, although complete spatial scans would be needed to achieve curvature comparisons by normalization of the data. In contrast, figure 3b shows comparison at 0.45% and 0.05% silane that exhibit a divergence between silicon atom spatial profiles and light scattering spatial profiles. Qualitatively, we conclude that spatial scans show a good spatial correlation of silicon atom signals with particle light scattering at mole fractions greater than 2% and less than 0.3% (except for the limiting behavior at 0.05%). The two processes of creating atoms by particle absorption and scattering laser light probably probe two different aspects of the particle distribution which may account for different spatial distributions at some mole fractions.

The experimental study of light scattering with short duration pulsed lasers does show a linear power dependence for particle light scattering done with 500.0 nm and silane concentrations of 5%. The corresponding power study has yet to be done in the ultraviolet, and a two-color, two-laser experiment is being planned to separate the effects of probe power from laser power. Tentatively, the demonstration [5] of a linear dependence of silicon atom signal on laser energy at powers above a threshold region is interpreted as particle absorption followed by atom separation. The linear dependence of the light scattering signal extends to low powers, which suggests that gross particle fragmentation into smaller particles is not important for this wavelength and power ranges that are ten times larger than the ultraviolet.

DISCUSSION

The concepts of particle nucleation and growth can be qualitatively applied to our observations. The composition of the particles is not yet known, but we will use a working hypothesis that electron and hydrogen atom impact serve to reduce the hydrogen content of a growing particle to yield Si_mH_n with $n/m < 1$. The hydrogen content of particles could well be reduced under some conditions into the range achieved by surface processes involved in amorphous film formation, although proof of particle composition awaits extension of our experiments.

The data in figure 2 that show spatial changes of particle light scattering with mole fraction of silane is compatible with general concepts of nucleation and growth. The lowest mole fraction of 0.05% has negligible light scattering at our current limit of detection, yet 0.01% silane shows particle scattering in the middle of the bulk plasma. This observation is compatible with achieving sufficient radical concentrations to allow nucleation and growth of particles in competiton with destruction by electron impact. A full spatial profile extending to the cathode is needed to examine the importance of electrode asymmetry in the nucleation process. It is possible that larger electron densities and energies at the cathode are responsible for initiating nucleation. Independent of the question of spatial origin of nucleation, figure 2 shows that successively increasing mole fractions of silane increase the light scattering intensity and spatial extent in the bulk plasma and also create a small scattering signal at the postulated location of the ion sheaths. If we recognize that the electron energy distribution may have high energy components at the ion sheath boundary, then a second process of nucleation and growth can easily be initiated at the ion sheath. The increasing dominance of particle light scattering at the sheath boundary with increasing silane mole fraction can be seen in the transition from 0.3% and 0.45%. Understanding this transition requires both

models and more information on number density versus size. The light scattering signal for particle sizes below the Mie particle limit (~ 40 nm) is a simple product of number density and the square of particle volume. The rapid change in scattered signal strength at the sheaths is most likely correlated with rapid growth to larger sized particles. If this is the case, then the sheath boundary becomes the most important location for particle growth because the particle destruction rate becomes much smaller than the radical creation and particle growth rates at suffciently large radical concentrations. Once a larger particle size distribution is established, it can then grow preferentially, as seen by the dramatic change in scattering intensity with flow rates (figure 1). We are beginning to formulate particle growth models based on these qualitative ideas and experimental results. New experiments to determine particle sizes, compositions, and number densities will be crucial for testing models.

The observation of silicon atom creation by particle absorption of pulsed laser radiation is potentially a very useful probe. While our current understanding of this linear energy absorption process needs more experimental refinement, we can propose two new applications for the process. One experiment will use the wavelength dependence of absorption to probe particle composition by using the efficiency of atom creation as a probe of electronic absorption. Another experiment could use the mechanism of particle absorpiton to probe particle size distributions and number density as a function of spatial location. This technique depends on the ultimate correctness of the conjectured absorption mechanism as being proportional to a product of particle number density times particle volume. This reasonable absorption mechanism, combined with the particle scattering efficiency as a product of number density and volume squared, can be used in simple ratios to identify spatial profiles of particle sizes and number density. Further experimental and theoretical work will be necesary to establish the ultimate promise of these proposed techniques.

ACKNOWLEDGMENT

We would like to thank the Office of Naval Research for support of this work. In addition, our co-workers G. D. Stein an G. Wong have been very helpful at earlier stages of this work.

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